

Optical Signature of Quantum Coherence in Fully Dark Exciton Condensates

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We predict that the collision of two fully dark exciton condensates produces bright interference fringes. So, quite surprisingly, the collision of coherent dark states makes light. This remarkable effect, which is many body in essence, comes from the composite boson nature of excitons, through the fermion exchanges they can have which transform dark states into bright states. The possibility of optically detecting quantum coherence in a regime where the system is hidden by its total darkness was up to now considered as hopeless.

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In this Letter we aim to write the final act of the half-century long drama on exciton Bose-Einstein condensation (BEC), namely, how to evidence the condensate wave function coherence by an optical mean when the condensate is fully dark. The *tour de force* we propose relies on the fact that carrier exchange between excitons couples dark to bright states. We use this many-body effect, exclusive to the composite boson nature of excitons, to here predict that the collision of two fully dark condensates must produce bright interference fringes that lead to a photoluminescence emission from an otherwise optically dark region (see Fig. 1). This effect constitutes the utmost evidence that the wave functions of the two colliding dark condensates are quantum coherent. To better grasp the importance of this challenge, let us first recall the previous acts of the exciton condensation drama.

The past.—The quest for BEC of semiconductor excitons—composite bosons (cobosons) made of one conduction electron and one valence hole—started in 1962 [1–3]. The critical density and temperature for BEC being easy to reach due to light carrier masses, excitons were for a long time thought to be the best candidate to experimentally produce this striking bosonic quantum effect. Yet, impressive progress in laser cooling has turned the tide and in 1995 the first ever BEC was realized in ⁸⁷Rb [4], ²³Na [5], and ⁷Li [6] atom gases.

As physicists were not understanding why exciton condensation eluded luminescence measurements, they turned to the polariton, which is a linear combination of one elementary boson, the photon, and one composite boson, the exciton. However, in microcavities where experiments were performed [7], the photon component in the polaritons that condense is very large and thus quite different from a genuine exciton. So, the problem of exciton BEC remained open.

The difficulty with exciton condensation is the complexity of the exciton physics, many aspects of which have to be

pieced together in order to possibly observe this quantum effect.

First, to avoid density collapse as well as condensate fragmentation into different momentum states [8,9], a repulsive interaction must exist between excitons. Since two excitons can bind into a biexciton molecule when their excitonic dipoles point in opposite directions, a way to avoid molecule formation is to use carriers located in spatially separate planes, as first proposed by Lozovik and Yudson [10]. This bilayer geometry is particularly attractive because “dipolar excitons” have a long lifetime that makes possible the study of cold exciton gases at thermodynamical equilibrium. Recent experiments [11–13] on exciton BEC have followed this idea [14].

Next, due to sizable spin-orbit splitting in the degenerate GaAs upper valence band and to quantum well confinement [15], the hole states with lowest energy are characterized by a quantum index commonly called “spin”

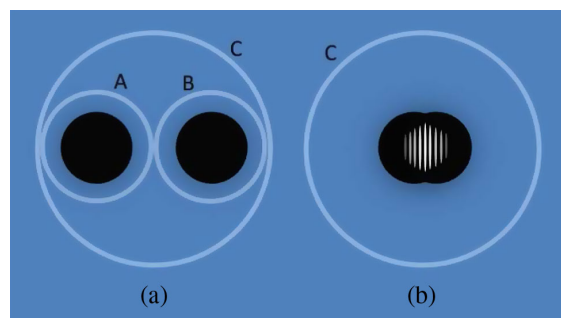


FIG. 1. (a) A dark condensate prepared in a large electrostatic *C* trap is split into two (dark circles) by turning on the *A* and *B* traps. When the (*A*, *B*) traps are turned off, the two dark condensates move toward the *C* trap center and interfere. (b) Schematic view of the effect we predict: bright interference fringes appear in the middle of the dark region, as a striking signature of coherence in the colliding fully dark condensates.

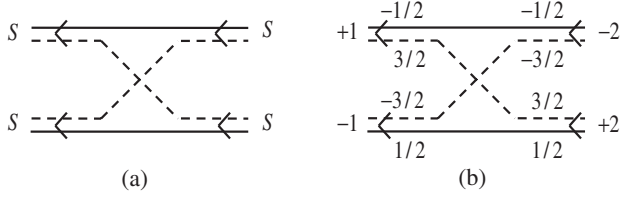


FIG. 2. Hole exchanges between two dark (± 2) and/or bright (± 1) excitons, visualized through Shiva diagrams [23,24]. Electrons are represented by solid lines, holes by dashed lines, and excitons by electron-hole double lines. Because exchanges conserve the carrier spin, excitons resulting from exchange between same $S = (\pm 1, \pm 2)$ excitons keep this S , as in (a). By contrast, exchange between dark excitons having opposite S leads to opposite- S bright excitons, as in (b), while exchange between a dark and a bright exciton would lead to the same dark and bright excitons [25].

$S_h = \pm 3/2$, while the conduction electrons are simply characterized by their spin $S_e = \pm 1/2$. Because electron-photon interaction conserves the genuine spin $s = \pm 1/2$, electron-hole pairs coupled to a photon, that are “bright,” are such that $S = S_e + S_h = \pm 1$, while for “dark” pairs not coupled to a photon, $S = \pm 2$ [16]. Since Coulomb interaction also conserves the genuine spin, bright excitons constructed on bright electron-hole pairs suffer interband Coulomb processes, while the dark excitons don’t. This (repulsive) interband Coulomb interaction pushes the bright excitons up in energy; so, the lowest states which are the ones that condense, are dark [17]. This allows associating exciton condensation with a darkening of the luminescence when the temperature decreases [18], provided the dark-bright energy splitting is small compared to the thermal energy, as for the GaAs bilayer, whose splitting is estimated to be $\sim 20 \mu\text{eV}$, that is, 10 times smaller than the thermal energy at 1 K.

Yet, the indisputable signature for condensation is the macroscopic coherence of the condensate wave function. It is clear that the dominant role played by the optically inactive states constitutes a severe constraint to experimental evidence, because the phase coherence of a fully dark condensate seemed optically unreachable. This impasse could only be unraveled through a deep understanding of the exciton composite nature and the interplay of their spin and orbital degrees of freedom. Excitons result from photon absorption; so, excitons are created in a bright state by construction. Yet, being made of indistinguishable fermions, carrier exchanges between excitons can transform two bright states into two dark states, and vice versa (see Fig. 2). As a result, (i) although excitons are created in a bright state, dark excitons do exist in the system, and (ii) through exchange interaction, unimportant in the very dilute limit, the dark condensate acquires a coherent bright component above a density threshold and turns “gray” [19]. Up to now, optical access to coherence has been possible in the gray regime: it is through the photoluminescence

emitted by its bright component that spatial and temporal coherences of the exciton condensate have been measured [20,21], and superfluidity observed from the formation of vortices [21,22].

Independently, Rapaport *et al.* [32] reported on a fully dark condensation of dipolar excitons, at densities well above the limit for which a gray condensate is energetically favorable, which is somewhat strange. It was argued that dipolar repulsion between excitons stabilizes the fully dark condensate, and inhibits a coherent introduction of bright excitons which prevents the dark condensate from turning gray. As no evidence of quantum coherence has been shown in this fully dark system, this point seems moot. The optical effect we here predict will not only allow probing BEC at density too low for the condensate to be gray, but also provide a way to prove that dark BEC does occur in the experiments reported in [32–34].

Physics of the predicted effect.—The interference pattern resulting from the collision of two condensates has been observed in the case of cold atoms [35–38]. The probability for detecting two elementary bosons located at $(\mathbf{R}_1, \mathbf{R}_2)$ is proportional to the two-boson spatial correlation function which has an oscillatory part in [39]

$$nn' \cos[(\mathbf{Q} - \mathbf{Q}') \cdot (\mathbf{R}_1 - \mathbf{R}_2)], \quad (1)$$

where (n, \mathbf{Q}) and (n', \mathbf{Q}') are the densities and momenta of the two condensates. This effect comes from the process of Fig. 3(a): a \mathbf{Q} boson and a \mathbf{Q}' boson are respectively observed at \mathbf{R}_1 and \mathbf{R}_2 . Being indistinguishable, the bosons which leave \mathbf{R}_1 and \mathbf{R}_2 can as well be \mathbf{Q}' and \mathbf{Q} bosons. This indistinguishability produces fringes with an oscillation characterized by the condensate momentum difference $(\mathbf{Q} - \mathbf{Q}')$. The nn' factor comes from the number of ways to choose the \mathbf{Q} and \mathbf{Q}' bosons among N and N' .

Because of their very small size, atoms behave as elementary bosons, and the only process that can happen is the one of Fig. 3(a) [35]. Indeed, fermion exchanges between cobosons occur within their relative-motion volume a^D , while the coboson center of mass is delocalized over the whole sample volume L^D , where a is the coboson Bohr radius and D the space dimension; so, these exchanges are controlled by the dimensionless many-body parameter

$$\eta = N \left(\frac{a}{L} \right)^D = na^D. \quad (2)$$

For atoms, η is close to zero while for excitons, whose size is much larger, η can be sizable; this explains why effects coming from carrier exchanges can be experimentally seen in excitonic systems.

We can add exchange to the diagram of Fig. 3(a) by connecting its two parts; we get the diagram of Fig. 3(b). If these exchanges occur between dark excitons having opposite spins, the excitons observed at \mathbf{R}_1 and \mathbf{R}_2 are

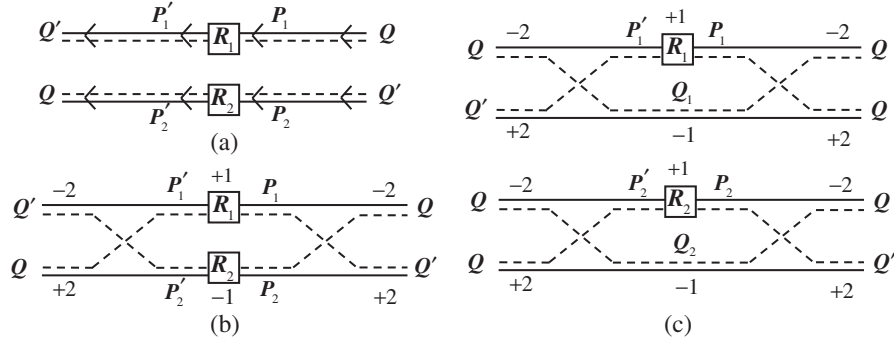


FIG. 3. (a) This diagram shows the process leading to Eq. (1), obtained for elementary bosons, or for cobosons in the absence of fermion exchange. (b) When exchange is introduced, the excitons observed at $(\mathbf{R}_1, \mathbf{R}_2)$ positions are bright, but this process does not lead to interference. (c) Disconnected diagram for hole exchanges leading to the $m = 1$ mode of Eq. (7). It involves three excitons from the \mathbf{Q} condensate and one exciton from the \mathbf{Q}' condensate. A similar exchange with two excitons \mathbf{Q} and two excitons \mathbf{Q}' leads to the $m = 2$ mode. The product of operators $B_{\mathbf{R}}^\dagger B_{\mathbf{R}}$ is visualized by an \mathbf{R} “box.” These operators located at \mathbf{R} are linked to operators for incoming and outgoing excitons having momenta \mathbf{P} and \mathbf{P}' through Eq. (8).

bright. However, this “connected” diagram [40] does not bring interference. The reason is that momentum conservation for the incoming $(\mathbf{P}_1, \mathbf{P}_2)$ and outgoing $(\mathbf{P}'_1, \mathbf{P}'_2)$ bright excitons

$$\mathbf{Q} + \mathbf{Q}' = \mathbf{P}'_1 + \mathbf{P}'_2 = \mathbf{P}_1 + \mathbf{P}_2 \quad (3)$$

leads to $\mathbf{P}_1 - \mathbf{P}'_1 = \mathbf{P}'_2 - \mathbf{P}_2$, which can take any value [25]. Interferences come from disconnected diagrams like the one of Fig. 3(c). Momentum conservation

$$\mathbf{Q} + \mathbf{Q}' = \mathbf{P}'_1 + \mathbf{Q}_1 = \mathbf{P}_2 + \mathbf{Q}_2 \quad (4a)$$

$$2\mathbf{Q} = \mathbf{P}_1 + \mathbf{Q}_1 = \mathbf{P}'_2 + \mathbf{Q}_2 \quad (4b)$$

then gives $\mathbf{Q} - \mathbf{Q}' = \mathbf{P}_1 - \mathbf{P}'_1 = \mathbf{P}'_2 - \mathbf{P}_2$. This process leads to bright excitons at $(\mathbf{R}_1, \mathbf{R}_2)$ due to the carrier exchanges it contains, and an oscillating correlation function in $\cos[(\mathbf{Q} - \mathbf{Q}') \cdot (\mathbf{R}_1 - \mathbf{R}_2)]$, which manifests as bright fringes [Fig. 1(b)].

Experimental proposal.—Indirect excitons formed in the GaAs bilayer provide a suitable platform to show the effect we predict. Recent experiments [34] have reached an exciton density as large as $5 \times 10^{10} \text{ cm}^{-2}$ in a trap with diameter $10 \mu\text{m}$ and potential depth 5 meV , which corresponds to a many-body parameter $\eta \simeq 0.2$ for an interlayer separation that leads to excitons having a Bohr radius $\simeq 20 \text{ nm}$.

The observation of photoluminescent interference fringes from dipolar exciton condensates, however, faces the very low optical activity of these excitons. It is then necessary to repeat the experiment a few million times in order to obtain a sufficient signal-to-noise ratio. When the relative phase of the two fully dark condensates is random, the resulting interference patterns will have bright fringes at different positions, and the averaging over these repeated experiments will blur the fringe pattern into a bright spot. This phase randomness is avoided when the two colliding

condensates come from the same source as proposed below, in analogy to the double-slit experiment.

What we suggest is somewhat similar to the procedure used for cold atoms [35]: we first load the C trap [Fig. 1(a)] with excitons at a temperature above the one for BEC, in order to ensure that the trap contains excitons. We then cool down the C trap until no light is emitted from it and we turn on the A and B traps to split the dark condensate into two, while keeping the excitons in the trap potential ground state. By suddenly removing the A and B traps, the C trap that encompasses the two condensates, exerts a force which pushes them toward its center with momenta \mathbf{Q} and $\mathbf{Q}' = -\mathbf{Q}$, where they interfere. Interference fringes are formed in the central region of the C trap: dark fringes, similar to the ones for atoms, exist but they cannot be seen. Bright fringes are also formed that can be optically detected. To produce bright fringes of width $\sim \mu\text{m}$, the C trap must be rather shallow compared to the depths of the A and B traps: indeed, a fringe width of $1 \mu\text{m}$ corresponds to an exciton momentum $\hbar/\mu\text{m}$, that is, a kinetic energy $\sim 3\mu\text{eV}$, which is provided by the C trap potential.

Mathematical support.—The interference pattern of two colliding condensates made of excitons with momenta $(\mathbf{Q}, \mathbf{Q}')$ and densities (n, n') , is obtained from the spatial correlation of two excitons located at $(\mathbf{R}_1, \mathbf{R}_2)$, these excitons having to be bright in order to be optically detected. The spatial correlation function for two bright excitons having circular polarizations (σ_1, σ_2) with $\sigma_i = \pm 1$, is given by

$$\langle B_{\mathbf{R}_1; \sigma_1}^\dagger B_{\mathbf{R}_2; \sigma_2}^\dagger B_{\mathbf{R}_2; \sigma_2} B_{\mathbf{R}_1; \sigma_1} \rangle, \quad (5)$$

the expectation value being taken in the two-colliding-condensate state $(N, \mathbf{Q}; N', \mathbf{Q}')$.

Case 1: If the two condensates were in the same bright state σ , Eq. (5) would give a spatial correlation that oscillates just as for elementary bosons [39] in Eq. (1),

provided $\sigma_1 = \sigma_2 = \sigma$. Indeed, when exchanges occur between same S excitons, this index does not change [see Fig. 2(a)].

Case 2: Likewise, carrier exchanges between same- S dark excitons do not produce bright excitons; so, the bright exciton destruction operator $B_{\mathbf{R};\sigma}$ acting on same- S dark condensates readily gives zero whatever σ : correlations do exist, but they cannot be optically detected.

Case 3: If one condensate contains (+2) excitons only and the other (-2) excitons only, carrier exchange between these opposite- S dark excitons produces bright excitons [Fig. 2(b)]. The two-bright-exciton correlation function differs from zero but no fringes are produced. Indeed, for bright fringes to appear, it is necessary to have a macroscopic amount of (+2) and (-2) excitons in one condensate at least, this condensate being either unpolarized $(B_2^\dagger)^{N_+}(B_{-2}^\dagger)^{N_-}|v\rangle$ or polarized $(g_2B_2^\dagger + g_{-2}B_{-2}^\dagger)^N|v\rangle$: the appearance of bright fringes definitely is a quite subtle many-body effect. Actually, through mean-field theory, it has been shown [17] that the dark exciton condensate is polarized with $|g_2| = |g_{-2}|$, thus making the formation of bright fringes *a priori* possible.

For a state made of two polarized dark condensates

$$|\psi_{N,N'}\rangle = (\mathcal{D}_{\mathbf{Q}}^\dagger)^N (\mathcal{D}_{\mathbf{Q}'}^\dagger)^{N'} |v\rangle \quad (6)$$

with $\mathcal{D}_{\mathbf{Q}}^\dagger = g_2B_{\mathbf{Q};2}^\dagger + g_{-2}B_{\mathbf{Q};-2}^\dagger$, we find that to the lowest order in density, the correlation function [Eq. (5)] oscillates with two modes,

$$|g_2g_{-2}|^4 \Lambda^4 \sum_{m=(1,2)} A_{\eta,\eta'}^{(m)} \cos[m(\mathbf{Q} - \mathbf{Q}') \cdot (\mathbf{R}_1 - \mathbf{R}_2)] \quad (7)$$

whatever (σ_1, σ_2) . The dimensionless coefficient Λ^2 contains two fermion-exchange scatterings, $A_{\eta,\eta'}^{(m)}$ being equal to $8\eta\eta'(\eta + \eta')^2$ for $m = 1$, and to $2(\eta\eta')^2$ for $m = 2$. Higher m modes do exist but they are of higher order in density.

To obtain this result, we used the composite boson many-body formalism [23,24], which allows handling fermion exchanges between cobosons in an exact way; its detailed derivation, including the calculation of Λ^2 in the case of GaAs bilayer, is given in the Supplemental Material [25]. The salient points about Eq. (7) are as follows:

(i) The g_2g_{-2} factor: it proves that the two types of dark excitons are necessary to produce a nonzero bright exciton correlation. This correlation is entirely due to fermion exchanges between (+2, -2) excitons, in the absence of Coulomb process; so, the associated exchange scattering that enters Λ^2 is dimensionless, unlike usual energylike interaction scatterings.

(ii) The $(\eta\eta')^m$ density dependence in the m mode and the $(\mathbf{Q} - \mathbf{Q}')$ difference: they show that the two condensates join together to produce oscillations, as for elementary bosons. Moreover, the existence of bright fringes supports a

polarized BEC because they require two types of dark excitons with same momentum.

(iii) The higher oscillatory modes $m = (2, 3, \dots)$, while elementary bosons only have the $m = 1$ mode [Eq. (1)].

Let us now go somewhat deeper into the calculation. The creation operator of a bright exciton σ located at \mathbf{R} is related to creation operators for excitons having a center-of-mass momentum \mathbf{P} through $B_{\mathbf{R};\sigma}^\dagger = \sum_{\mathbf{P}} B_{\mathbf{P};\sigma}^\dagger \langle \mathbf{P} | \mathbf{R} \rangle$ with $\langle \mathbf{P} | \mathbf{R} \rangle = e^{-i\mathbf{P}\cdot\mathbf{R}}/L^{D/2}$; so,

$$B_{\mathbf{R}_1;\sigma}^\dagger B_{\mathbf{R}_1;\sigma} = \frac{1}{L^D} \sum_{\mathbf{P}'_1, \mathbf{P}_1} e^{i\mathbf{R}_1 \cdot (\mathbf{P}'_1 - \mathbf{P}_1)} B_{\mathbf{P}'_1;\sigma}^\dagger B_{\mathbf{P}_1;\sigma} \quad (8)$$

We visualize this product of operators by a \mathbf{R}_1 box with an incoming bright exciton \mathbf{P}_1 and an outgoing bright exciton \mathbf{P}'_1 (see Fig. 3). When the excitons making the condensates are all dark as in Eq. (6), the exciton composite nature must enter into play through fermion exchanges as in Fig. 2(b), in order for the $(\mathbf{P}', \mathbf{P})$ excitons to be bright. Starting from the diagram of Fig. 3(a), the simplest way is to have the observed $(\mathbf{R}_1, \mathbf{R}_2)$ bright excitons resulting from a hole exchange between a dark exciton (+2) from the \mathbf{Q}' condensate and a dark exciton (-2) from the \mathbf{Q} condensate. This process, shown in Fig. 3(b), could be optically detected for $\sigma_1 = -\sigma_2$, but not for $\sigma_1 = \sigma_2$. Momentum conservation given in Eq. (3) produces terms in $e^{i(\mathbf{R}_1 - \mathbf{R}_2) \cdot (\mathbf{P}'_1 - \mathbf{P}_1)}$ through Eq. (8), but does not enforce $(\mathbf{P}'_1 - \mathbf{P}_1)$ to be constant. So, it does not lead to fringes.

The more complex process of Fig. 3(c) also contains carrier exchanges between (+2, -2) excitons, as necessary to have bright excitons at $(\mathbf{R}_1, \mathbf{R}_2)$, but momentum conservation given in Eq. (4) now imposes $\mathbf{P}_1 - \mathbf{P}'_1 = \mathbf{Q} - \mathbf{Q}'$, which leads to a constant phase in Eq. (8), and similarly $\mathbf{P}_2 - \mathbf{P}'_2 = \mathbf{Q}' - \mathbf{Q}$. So, we end with a $\cos[(\mathbf{Q} - \mathbf{Q}') \cdot (\mathbf{R}_1 - \mathbf{R}_2)]$ term by interchanging $(\mathbf{R}_1, \mathbf{R}_2)$, that is, a $m = 1$ mode. Since this process involves three \mathbf{Q} excitons and one \mathbf{Q}' exciton, it must appear with a $\eta^3\eta'$ density dependence. Moreover, since it involves two pairs of excitons (+2, -2) on both sides, taken from the polarized $|\psi_{N,N'}\rangle$ condensate, it has to contain a $|g_2g_{-2}|^4$ factor. This, and the other two processes involving one \mathbf{Q} exciton and three \mathbf{Q}' excitons, and two pairs of $(\mathbf{Q}, \mathbf{Q}')$ excitons, yield the $m = 1$ term in Eq. (7). Note that for the process of Fig. 3(c) to produce an oscillation, the \mathbf{Q} condensate must contain (+2) and (-2) excitons, while the \mathbf{Q}' condensate can contain (-2) excitons only.

Another process, similar to the one of Fig. 3(c), but with two \mathbf{Q} excitons and two \mathbf{Q}' excitons on the same side, instead of (\mathbf{Q}, \mathbf{Q}) and $(\mathbf{Q}, \mathbf{Q}')$, also has a $|g_2g_{-2}|^4$ factor, but a density dependence in $(\eta\eta')^2$. Momentum conservation now imposes $2\mathbf{Q}' = \mathbf{P}'_1 + \mathbf{Q}_1$ and $\mathbf{P}_1 + \mathbf{Q}_1 = 2\mathbf{Q}$, which give $2(\mathbf{Q} - \mathbf{Q}') = \mathbf{P}_1 - \mathbf{P}'_1 = \mathbf{P}'_2 - \mathbf{P}_2$; so, this process brings the $m = 2$ term of Eq. (7). For the $m = 2$ mode

to appear, the two dark condensates have to both contain (+2) and (-2) excitons.

Discussion.—To keep the relative phase of the two condensates fixed for each repeated experiment, we must allow the particle numbers of the two condensates to vary, since the system phase and the particle number are conjugate variables. Let us for simplicity consider that excitons are elementary bosons interacting through an effective two-body potential ξ^{eff} . Because the condensate energy depends on the number of particles it contains, the time evolution operator e^{-iHt} is going to produce different phases to the two evolving condensates having different particle numbers. Mean-field calculation [41] actually shows that the diffusion (root-mean-square deviation) of the relative phase increases with time t as $\sqrt{N}\xi^{\text{eff}}t$, the \sqrt{N} dependence coming from the fluctuation in the N -particle binomial distribution of the two traps. So, if the time lapse between the condensate splitting and the detection is short, the interference pattern will stay the same in each experiment.

To conclude, we propose an optical way to probe quantum coherence in an excitonic system hidden by its darkness. Such a signature seemed at first hopeless. The effect we here propose is based on the fact that two opposite-spin dark excitons transform into bright excitons through carrier exchange scatterings, that are dimensionless. The bright fringes we predict from the collision of two fully dark exciton condensates constitute the utmost evidence of coherence in these hidden states.

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